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RADIOCHEMICAL STUDIES FOR THE NUCLEAR CHEMICAL MINING OF COPPER

Donald D. Jackson

February 19, 1976

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RADIOCHEMICAL STUDIES FOR THE NUCLEAR CHEMICAL MINING OF COPPER

Abstract

Experiments were performed to study the long-term reactions of radioactive materials that would be produced in the nuclear chemical mining of chalcopyrite copper ore.

These reactions were carried out in autoclaves for up to eight months at 2.76 MPa of oxygen and 363 K. Dissolving rates of radioactive glass in copper-leaching product solutions were determined by the gamma-counting of solution aliquots. The glass decomposition rate was linear at approximately 1×10^{-10} kg/s·m². A radioactive tracer technique was used to study the extent of sorption of seven fission product nuclides on ore

and decomposition products during leaching as functions of ore particle size, solution composition, pH, and liquid/solid ratio. The distribution of radionuclides between solid and liquid phases was determined. In general, the solids continue to concentrate the radioactivity from the liquid for long periods of time, as the ore and gangue surfaces change in the complex chemical and physical processes of leaching. Solid phases in a nuclear chemical mine would therefore effectively decontaminate the leach liquor. Leaching time and pH are variables that could be used to control the level of activity in solution.

Introduction

The *in situ* mining of copper from large, deep, primary sulfide ore bodies that have been fragmented by nuclear explosives, called nuclear chemical mining, has been suggested as an alternative to conventional methods of copper mining. Extensive investigations and process develop-

ment have been carried out at the Lawrence Livermore Laboratory to establish the technical and economical feasibility of such a mining method.¹⁻¹⁵

In nuclear chemical mining, a large, rubble-filled cavity, or chimney, would be formed by a nuclear

explosion and flooded with water. Oxygen would be pumped in and circulated through the rubble volume. Under these conditions, the sulfide minerals pyrite and chalcopyrite are oxidized in exothermic reactions to produce water-soluble copper sulfate, sulfuric acid, and mixed solid residue phases, including sulfur and jarosites, which are hydrous iron sulfates. The kinetics of these leaching processes, including the important reactions (1) through (3) have been studied by Braun *et al.*⁷

After the leach solution has reached its optimum composition, it would be pumped to an above-ground processing plant for the recovery of copper by solvent extraction and electrowinning processes.

The purpose of the present work is to investigate the possible radioactive contamination in such a mining operation from the fission and activation products of the nuclear explo-

sive. After the ore body has been shattered, some of these radio-nuclides will be distributed on the surfaces of ore fragments, and some will be incorporated in rock that has been melted.¹⁶⁻²²

The leach solutions used to extract copper can also leach and transport radioactive trace elements from the chimney. These trace elements undergo complex chemical and physical reactions that are not well understood. Some fraction of the radioactivity that is dissolved from the chimney rubble remains in solution. The remainder is sorbed by glass, by unmelted rock, and by their decomposition products. Such processes as precipitation, sorption, and rock dissolution determine the nuclide availability, or the amounts of fission products that can be transported in solution and suspension.

Information on this availability is an important first-consideration in evaluating the possibilities of transmission of radioactivity from the chimney to the processing plant or into aquifers through which ground water might be contaminated. The interactions of radioactivity with ground water systems have been considered in detail by Levy.²³ An estimation of the magnitudes of all radioactivity problems associated with nuclear copper mining has been made by Tewes *et al.*⁹

In the present work, two general types of studies were undertaken, both under conditions of temperature, pressure and oxygen flow similar to those expected in a process of this type. Experiments were done to determine the leaching rates of radionuclides from actual melt glasses. The method of computing these rates is given in Appendix A. In other experiments, samples of ore were leached with solutions containing tracer radionuclides to obtain data on the distribution of various species of interest between solid and liquid phases. The variables explored in the ore leaching experiments were time, particle size, solution composition, and liquid/solid ratio.

The parameters determined were distribution coefficients, as defined by Eqs. (4) through (8):

Mass distribution coefficient

$$K_{dm} = A_s / A_l \quad (4)$$

Area distribution coefficient

$$K_{da} = R \cdot K_{dm} \quad (5)$$

Solid radioactivity

$$A_s = \text{disintegrations/Mg} \cdot \text{s} \quad (6)$$

Liquid radioactivity

$$A_l = \text{disintegrations/m}^3 \cdot \text{s} \quad (7)$$

Solid mass/surface area ratio

$$R = \text{kg/m}^2 \quad (8)$$

Thus, K_d may be regarded as a figure of merit. Species with high values for K_d remain largely on solid phases and could be removed from the chimney only when these solids are particulates in suspension. Radionuclides with low K_d values would contaminate the leaching solutions, and if present in sufficiently high concentrations, would require special treatment.

Most K_d values reported in the literature correspond to K_{dm} . They are usually measured for very finely divided materials, where the entire particle appears to participate in the sorption process and the surface area involved is poorly defined. However, for larger particles, the sorptive capacity is expected to correlate better with the macroscopic surface area and, hence, K_{da} is a more significant parameter.

Experimental

EQUIPMENT

Six experiments (designated A through F) were performed in separate

autoclaves. These autoclaves were charged with various combinations of leaching solutions, chalcopyrite ore, and radioactive melt glass. The

characteristics of the autoclaves, their contents, and the physical conditions are summarized in Table 1.

A schematic diagram of a typical autoclave assembly is given in Fig. 1. Figure 2 shows a pair of autoclaves enclosed to contain radioactivity. All autoclave parts exposed to the fluids were titanium or AISI 316 stainless steel, except for valve seals and oxygen inlet tubes, which were made of polytetrafluoroethylene.

Temperatures were measured by iron-constantan thermocouples in wells inside the autoclave. Temperature was controlled by meter-relay-type controllers and temperatures were usually maintained within 1 to 2 K of the set point. An oven-type reference junction was used and the temperature for each experiment was printed by a data acquisition system, usually at hourly intervals.

COPPER ORE

Samples of primary sulfide ore from the San Manuel Mine (Magma Copper Co.,^{*} Arizona) were used for these experiments. The ore was crushed, screened, and washed with distilled water to remove surface dust. Two size ranges were used: 6.35 to 9.52 mm (1/4 to 3/8 in.) and 38.1 to 76.2 mm (1 1/2 to 3 in.).

RADIOACTIVE MELT GLASS

Since no nuclear experiments have been carried out in primary sulfide ore bodies, glassy radioactive rock

^{*}Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Energy Research & Development Administration to the exclusion of others that may be suitable.

Table 1. Experimental conditions.

Experiment	A	B	C	D	E	F
Autoclave material	AISI 316	AISI 316	AISI 316	AISI 316	Titanium	Titanium
Autoclave volume (l)	0.300	0.300	3.785	3.785	0.300	0.300
Solution type	leach solution	leach solution	leach solution	water	leach solution	leach solution
Solution weight (g)	105.1	173.5	2332	1801	105.0	171.1
Ore weight (g)	165.1	—	475.2	1801	165.0	—
Ore size (mm)	6.35 to 9.52	—	38.1 to 76.2	38.1 to 76.2	6.35 to 9.52	—
Melt glass weight (g)	8.3	13.5	—	—	—	13.5
Melt glass size (mm)	0.59 to 1.19	0.59 to 1.19	—	—	—	0.59 to 1.19
Liquid/solid ratio	0.61	12.67	4.91	1.00	0.64	12.67
Reaction time (d)	48.1	47.0	91.8	106.4	255.6	250.6
Oxygen	2.76 NPa, 4.0 cm ³ /s flow					
Temperature	363 K					

Fig. 1. Typical autoclave assembly.

Fig. 2. Enclosed autoclave for containing radiation.

of the desired composition was not available for leaching and sorption studies. The material used to simulate its behavior was radioactive glass recovered from a nuclear experiment in Nevada tuff. Drill cores from several locations in the melt region of this experiment were combined and washed with water and acetone to remove the clay matrix in which they were recovered. The residue was composed almost entirely of fragments of hard, black glass. These were crushed and screened into six grain-size fractions. Samples were analyzed for gamma activity, chemical composition, and density. The melt glass used in these experi-

ments was from the 0.59 to 1.19-mm fraction. Its density was 2.38, and its chemical composition is shown in Table 2. The radiochemical composition is given in Table 3. The radionuclides that were leached from the glass and that could be measured in these experiments were ^{144}Ce , ^{152}Eu , ^{154}Eu , and ^{155}Eu . The activity of the refractory nuclides decreased slightly but consistently with decreasing particle diameter. This change was about 12% between 0.1-mm and 1-mm diameter glass grains and is attributed to differences in crushing resistance of glass and tuff impurities and therefore to progressive dilution of glass by softer material of slightly different radiochemical composition.

Table 2. Chemical compositions of melt glass and chalcopyrite ore.

Component	wt% in glass	wt% in ore
Cu	—	0.7
Fe	1.5	3.2
S	—	2.2
SiO_2	72.3	66.9
Na_2O	4.3	1.5
K_2O	4.3	2.5
CaO	1.9	0.9
MgO	0.47	0.5
Al_2O_3	16.1	15.5
CO_2	—	0.4
TiO_2	0.33	—
Ba	0.05	—
Sr	0.004	—

LEACHING SOLUTIONS

The leaching solutions for experiments A, B, C, E, and F had initially the composition described in Table 4, an ionic composition resulting typically from the long-term leaching of the chalcopyrite ore used. Additionally, the pH of this solution used in experiment E was adjusted to 1.0 at 294 K by the addition of sulfuric acid. By using such leach solution compositions, it was anticipated that the time required to reach steady-state sorption conditions could be reduced. Experiment D was begun with

Table 3. Radiochemical composition of melt glass.

Gamma activity ratios at reference time for experiments F and F							
	$^{137}\text{Cs}/^{134}\text{Cs}$	$^{90}\text{Sr}/^{137}\text{Sr}$	$^{137}\text{Cs}/^{134}\text{Cs}$	$^{152}\text{Eu}/^{154}\text{Ce}$	$^{152}\text{Eu}/^{144}\text{Ce}$	$^{155}\text{Eu}/^{144}\text{Ce}$	$^{182}\text{Tl}/^{144}\text{Ce}$
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	
0.100 ± 0.005	0.0000	0.0000	1.100	1.100	0.533	0.00269	

distilled water, and its composition was allowed to change naturally.

Tracer radionuclides were added to each leaching solution to permit K_d measurements and to allow the total quantities of species dissolved from glass samples to be calculated. The tracer species used were ^{85}Sr , ^{88}Y , ^{103}Ru , ^{124}Sb , ^{134}Cs , ^{141}Ce , and ^{152}Eu .

There was a physical restriction on the relative amounts of ore, solution, and glass that could be used experimentally. In an actual mining operation, these materials are estimated to be in the mass ratio 38/3/1, respectively. At a 75% volume filling, an experiment in a 300 ml autoclave would require about 430 g ore, 33 g solution, and 11 g glass. This was an inconveniently small amount of liquid for these experiments. The ratios actually used are given in Table 1.

A few analyses of the compositions of leaching product solutions were

Table 4. Composition of stock leaching solution.

Species	Concentration (mg/ml)
Cu ⁺⁺	4
Mg ⁺⁺	2
K ⁺	0.9
Al ⁺⁺⁺	.8
Ca ⁺⁺	.5
Mn ⁺⁺	.2
Fe ⁺⁺⁺	.1
Zn ⁺⁺	.05
Ni ⁺⁺	.03
Na ⁺	.02
Co ⁺⁺	0.02
SO ₄ ⁼	25
NO ₃ ⁻	0.2
Cl ⁻	0.02
pH	adjusted to 1.7 with concentrated H ₂ SO ₄

made during the course of the experiments. These are recorded in Table 5.

Table 5. Chemical compositions of leach solutions (mg/ml).

Experiment	Reaction time (d)	Cu	Mg	Ca	Fe	Na	K	Si	Al	SO ₄ ²⁻
B	6.987	1.54	1.77	0.35	0.25					
	18.782	1.68	1.78	.24	.15					
	25.832	1.71	1.85	.28	.13					
	32.826	1.84	1.88	.27	.10					
	40.193	1.48	1.56	.23	.04					
	46.982	1.23	1.25	.24	.03					
C ^a	94.646	2.55	2.36	.54	.04	0.04	0.56	0.31	0.42	21.8
F	-1.179	1.66	1.80	.46	.10					
	2.990	1.74	1.89	.47	.11					
	6.045	1.78	1.90	.45	.09					
	9.649	1.73	1.86	.47	.06					
	16.794	1.82	1.88	.45	.04					
	22.629	1.81	1.92	0.41	0.04					
	81.365							.135		
	88.365							.137		
	95.365							0.134		

^aDecanted cool solution.

SAMPLING AND ANALYSIS

Samples of radioactive leach solutions were removed and returned to the autoclave periodically through the attached valves and sample bottles without cooling the experiment or releasing the pressure. After an initial period of rapidly changing K_d values, samples were usually taken at weekly intervals.

Each sample was drained into a polyethylene bottle and weighed. A

pH measurement was taken, and the sample reweighed. An aliquot was then removed for filtration. The filter was a polycarbonate membrane with a pore size of 0.1 μ m and supported on a 90-mm polyethylene Buechner funnel. Sorption of radioactivity by filtration equipment was in all cases found to be negligible. The filtrate was collected in a counting bottle and analyzed for gamma activity. Counted samples ranged from 3 to 9 g.

Table 6. Radioactivity of filter residues from experiment E (disintegrations/s·mg).

Time (d)	¹⁰³ Ru	¹²⁴ Sb	¹⁴¹ Ce	¹⁵² Eu	¹³⁴ Cs	⁸⁸ Y	⁸⁵ Sr
9.841	27.83	147.77	85.98	0.5118	3.303	1.5838	102.42
40.698	12.90	50.58	41.58	.3815	1.572	—	55.05
60.858	25.20	79.05	70.88	.3315	1.780	0.4552	99.93
68.732	3.62	11.64	10.83	.0555	0.282	.1027	15.06
74.880	13.91	44.47	37.33	.0881	1.257	.2288	53.85
82.870	38.35	121.60	105.27	.1467	3.657	.5220	138.38
89.696	14.10	42.83	38.97	.1207	1.364	.2137	52.12
110.608	6.06	16.73	15.27	—	0.702	.1068	22.90
117.611	14.11	40.42	35.00	—	1.577	.1998	54.23
124.604	7.02	19.45	18.08	—	0.929	.1000	26.70
131.625	8.46	29.32	23.28	.0519	1.286	.1414	30.42
138.625	7.66	22.70	21.63	—	1.323	.1111	30.70
145.644	2.78	7.80	6.24	—	0.649	.0850	9.45
152.644	11.70	36.20	28.62	.0664	2.162	.2562	47.40
159.678	4.09	16.20	12.29	.0294	0.697	.0449	14.99
166.594	11.00	33.72	29.74	.0642	1.089	.0683	45.93
173.639	15.33	45.61	40.56	.0569	1.297	.1073	63.43
180.604	18.04	58.75	56.31	.0394	2.829	0.1962	70.28
187.604	138.8	405.2	340.6	.7401	49.69	3.086	558.8
194.604	9.93	33.41	32.31	.0525	1.617	—	41.49
201.604	2.79	4.96	7.59	.0218	0.922	—	7.36
211.907	689.3	28.19	26.76	—	0.670	—	37.14
215.604	8.97	22.44	18.14	—	1.293	—	28.84
222.584	—	19.09	29.02	—	2.616	—	18.55
228.625	5.84	16.07	16.27	.0245	0.8811	0.1477	21.27
236.644	—	7.66	12.54	0.0131	0.7835	—	8.24
243.644	—	16.64	—	—	6.97	—	25.63
250.625	—	279.83	—	—	34.35	—	436.33
255.625	—	12.16	22.15	—	1.21	—	17.65

Usually, the quantity of filter residue was very small. In experiment E, there was often enough suspended material to give a filter residue that could be weighed and counted with reasonable accuracy (Table 6).

Nonradioactive stock leaching solution was added to the unused sample to replace the counting aliquot and any liquid lost by transpiration in the oxygen stream. This volume was then returned to the autoclave.

Results

Dissolving rates for the radioactive melt glass and mass distribution coefficients K_{dm} for the six experiments are tabulated in Appendix B. Each table of K_{dm} values gives the conversion factor for computing the area distribution coefficient K_{da} . Composite plots of K_{da} vs time for each radionuclide on glass and on ore are given in Appendix C. The data were plotted this way to illustrate the effects of solution composition. Some observations for the individual experiments are noted below.

EXPERIMENT A

In this experiment, both glass and ore were leached. The glass was first placed in the bottom of the autoclave and the ore piled on top, simulating the situation in a nuclear chimney. The K_d values are therefore for total solids. However, the mass and K_d ratios are such that sorption

by the glass can be neglected.

In subsequent experiments, ore leaching and glass dissolving were done separately because the two types of studies involve very different levels of radioactivity.

EXPERIMENT C

After the completion of experiment C, the autoclave was cooled to room temperature at continued oxygen pressure and flow. Sampling indicated that this treatment had a negligible effect on K_d values, except perhaps for the reappearance of ruthenium in the solution at sufficient activity to be measured.

The residues formed in this experiment are illustrated in Figs. 3-7. They are typical of products of the other ore leaching experiments. A brief study was made of the distribution of gamma activity in these residue phases, and their components were identified by x-ray diffraction.

Fig. 3. Autoclave and contents after Experiment C.

Fig. 5. Sulfur deposits on flange after Experiment C.

Fig. 4. Autoclave flange assembly after Experiment C.

Fig. 6. Deposits of anhydrite and jarosite on oxygen inlet tube after Experiment C.

Gamma activity was studied by examining selected individual samples, but no attempt was made to homogenize the residues to achieve a complete gamma activity balance. These data are summarized in Table 7. Some pieces of ore had disintegrated into smaller fragments while others remained intact. There was a considerable amount of loose material,

Fig. 7. Anhydrite deposits on sampling tube after Experiment C.

Table 7. Distribution of activity at the end of experiment C.

Type of residue	Mass (g)	Percentage of original radionuclides found in residue							
		^{137}Cs	^{134}Cs	^{141}Ce	^{152}Eu	^{138}Ce	^{88}Y	^{89}Sr	
Solution, 0.1 M filtrate	195,0039	0.27	—	—	5.27	3.46	6.64	—	
Solution, 1.0 M filtrate	477,1680	0.73	0.07	2.08	27.81	6.37	21.66	0.58	
Solution, decanted	1614,8834	—	—	—	51.30	28.84	60.86	—	
Filter residue, 0.1 M	0.0082	—	—	—	0.01	—	0.01	—	
Gypsum and anhydrite crystals from outside sampling tube	1759	—	—	0.13	0.50	—	0.19	—	
Gypsum and anhydrite crystals from inside oxygen inlet tube	1834	—	0.23	1.77	11.24	0.08	5.55	0.02	
Sulfur from underside of autoclave flange	(lost)	—	—	—	—	—	—	—	
Jarosite coating on oxygen inlet tube	0,0233	1.53	1.35	1.42	0.15	0.01	0.05	1.56	
Fine (<1 mm) precipitate of jarosite, muscovite and some gypsum from autoclave	38,8611	51.73	35.22	36.96	6.82	13.53	—	33.14	
Coarse (>1 mm) precipitate and gangue	32,6029	—	11.56	—	2.25	10.54	—	8.47	
Large ore fragments and coatings	428,3842	—	4.14	—	9.74	57.15	—	31.96	
Total activity removed during sampling		0.14	0.60	1.30	3.65	2.43	4.10	0.68	
Total activity accounted for		43.90	53.16	43.66	112.74	122.41	99.06	76.41	

principally jarosites [nominally $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$ but variable in composition] covering the ore and the bottom of the autoclave. Much of the activity had been absorbed by this residue.

There were well-formed crystals of anhydrite (CaSO_4) and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) on the sampling tube and inside the oxygen inlet tube. These were relatively enriched in yttrium and europium but not in strontium, as might have been expected. Strontium behaved chemically much like antimony and ruthenium. Most of the anhydrite crystals probably formed late in the experiment, when the available nuclides were largely those with low K_d values for other phases. There was a bright yellow coating of jarosites on the outside of this oxygen

inlet tube. The titanium thermocouple well was similarly coated. Sulfur had been deposited on the underside of the top flange. The activity was low in this phase with cesium being the tracer present at highest activity.

EXPERIMENT D

After the experiment, a large quantity of gypsum and anhydrite crystals were noted. No loose, yellow precipitate layer was found in the bottom of the autoclave, as in experiment C. Instead, the ore fragments had a reddish coating.

Some species showed unusual behavior in this experiment, where the fluid was initially water. There were high early K_d values for cerium, yttrium, and europium, which then

declined rapidly, perhaps because of falling pH as sulfuric acid was generated in the leaching process. In this experiment only, strontium had a low K_d at early times. This may be associated with the concentration of calcium, with which it would be expected to co-precipitate.

At the end of this experiment, the solution was cooled to ambient temperature and resampled. The results of this treatment can be seen in the final data for this experiment and vary from little change for europium and yttrium to moderate changes in both directions

Fig. 8. The pH history of Experiment E.

for other nuclides. Values for antimony and ruthenium could again be measured at this time, suggesting that cooling had caused a substantial desorption.

EXPERIMENT E

The starting time for this experiment refers to the beginning of heating. Relative to this, the actual liquid-solid contact time at 298 K and the beginning of pressurized oxygen flow were at -2.877 d. A sample of the cool solution was taken at -2.125 d, as shown in Appendixes B and C.

An attempt was made to maintain the pH at approximately 1.0 by periodically adding concentrated H_2SO_4 . The difficulty in maintaining this pH is illustrated by the pH vs time history in Fig. 8. The variability occurred because the rate of acid consumption was not constant during the first three months. After this, there was little change, and acid additions were discontinued.

The K_d values showed wider fluctuations than in other experiments, probably reflecting pH changes. They changed more regularly after acid was no longer added.

The curves for all species show discontinuities on or after the seventy-fifth day of the experiment. The cause of this is not definitely known, but it is probably associated with a temperature excursion to 366 K (93°C) for about 24 h on days 71 to 72. The effect was transitory for antimony, europium, cesium, and yttrium, while ruthenium, cerium, and strontium show lasting offsets.

EXPERIMENT F

The starting time for this experiment refers to the beginning of heating. Relative to this, the actual liquid-solid contact time at 298 K and the beginning of pressurized oxygen flow were at -2.919 d. A sample of the cool solution was taken at -1.179 d, as shown in Appendixes B and C.

Discussion

SORPTION EXPERIMENTS

The sorption of radionuclides on minerals and soils has been studied by many investigators. Rancon²⁴ dis-

cussed some of the problems of measuring K_d values on natural materials, including the utility of mass and surface area K_d functions in different particle size ranges. Jenne

and Wahlberg²⁵ pointed out the relation of K_d to equilibria in real systems through the mass-action selectivity coefficient. This is in some respects a more generally useful function in that it involves not only the sorptive capacity of a solid phase, but the activities of the several ions that compete for available sites.

The copper ore used in these experiments is a heterogeneous material, consisting predominantly of quartz and sericite, with minor phases of feldspar, chlorite, biotite, calcite, chalcopyrite, pyrite, and hematite.⁷ The sorptive properties of these component minerals for radionuclides have not been studied under the conditions employed here. However, the rates of such sorption reactions seem to be of less importance than the fact that this is a reactive system.

The ore is decomposed by the oxidizing acidic solution, and the compositions of the solid and liquid phases change continuously. Not only do additional mineral surfaces become accessible as the leaching proceeds, but precipitates are produced that provide fresh sites at which sorption can take place.

The K_d vs time functions observed for the various species are composites of interactions with a changing mineral assembly and cannot be

described in terms of a simple model. It is desirable to describe the behavior of the radionuclides by K_d expressions in which the simple gross initial mass or surface areas were replaced by summations reflecting the existing mineral phases and their particle size distributions as a function of time. Adequate and continuous characterization of the physical or chemical states of the solid and liquid phases was not possible in these preliminary experiments. Such studies should be carried out, however, when the conditions for an actual mining experiment are precisely defined.

The most useful result of the present study was that the sorption reactions did not quickly come to a steady state. In most cases, they continued as long as the experiment was carried out. The activities of species, notably ruthenium and antimony, sometimes became so low in solution that they could no longer be measured. The longer the leaching process is allowed to continue in an actual mine, therefore, the less the solution will be contaminated by radioactivity.

Although all the radioactive species were involved in these rate processes, they showed characteristic behavior as to relative magnitudes and rates of change of K_d . In general, the order of decreasing

magnitude of K_d values for the species was antimony, strontium, ruthenium, cerium, cesium, europium, and yttrium. Europium and yttrium consistently showed low K_d values and a remarkable similarity in behavior. They showed most nearly consistent behavior in different ore experiments. They were strongly sorbed by gypsum and anhydrite, but not by jarosites. This very likely accounts for the fact that their K_d values were not greatly different on glass than on ore. For other species, the glass surfaces sorbed activity much less strongly than the ore phases.

In experiment E, an attempt was made to determine to what extent the radiochemical composition of the precipitated material in suspension reflected the changing composition of the solution. The filter residues were weighed and counted, and the activity/mass ratios given in Table 7 were determined. There is considerable scatter in the data, probably a result of the sampling procedure because the particle sizes of suspended particulates transferred to the filters were not always the same. However, specific activities for all species appear to have decreased with time, as expected of continuing precipitation in a solution of decreasing activity. The activities of species with high K_d values increased with time in the suspended

material relative to those with low K_d values.

Despite the irregular course of K_d vs time curves, the composite plots show that ultimately the effect of lower pH in experiment E was to reduce the amount of activity sorbed, for ruthenium, antimony, cerium, cesium, and strontium. No such pH effect was observed for europium and yttrium. On this basis, then, it appears less desirable to operate an *in situ* leaching process at a low pH by the addition of sulfuric acid.

This pH effect also implies that in so far as the transport of activity by ground water is concerned, as leaching solutions move from the vicinity of the chimney and acid and oxygen are consumed by reaction, the extent of sorption should increase. This is supported by experiment D, where the leach solution was initially water and K_d values increased more rapidly at early times than in other experiments.

In experiment C, the virtual absence of activity that was condensed on cooler autoclave surfaces or in the enclosure, suggests that volatilization of radioactive species did not take place. However, the accuracy of accounting for total activities recovered in this experiment, as shown in Table 7, was not sufficient to confirm this.

The liquid/solid ratios in Table 1 were higher than the value of about 0.08 that might be expected in an actual nuclear mine.² This should delay sorption in reactions involving saturations of the solution and precipitation of a new phase such as silica or anhydrite. However, such effects were not definitely observed.

GLASS DISSOLVING EXPERIMENTS

Glasses do not have definite solubilities in water. They have only decomposition rates, which continue until the materials are entirely consumed.

There is extensive literature on these reactions.^{26,27} However, the mechanisms involved are complex and are sensitive to slight variations in physical and chemical composition. The result is that there is no general method of predicting corrosion rates from theoretical considerations. The determination of glass durability remains largely an empirical study, and only the most general correlations can be made.

Hydration of a glass surface is the first stage of decomposition and is followed by extraction of the mobile alkali ions near the surface. If the silica content is high enough to exist in the glass as a continuous network, a siliceous corrosion layer forms on the surface. Further reac-

tions then involve exchange with and diffusion through this layer.

Industrial glasses are usually made with compositions and processes to produce homogeneous articles. Glasses created by melting rock under uncontrolled conditions are likely to contain inhomogeneities on a sub-microscopic scale.^{16,27} Ions associated with separated minor phases can leach at rates different from those for the bulk of the glass. The exhaustion of such material from near the surface can decrease the apparent dissolving rates of some species.

Many leaching experiments with radioactive glasses and silicate minerals¹¹ have been carried out. The extraction of activity from samples of debris recovered from nuclear experiments has been studied.²⁸⁻³³ As expected, the rate of removal of radioactivity increases with increasing temperature, decreasing particle size, and in very alkaline or acidic solutions.

The radioactive glass used in this study has higher silica and alkali contents than the ore and, presumably, than glass produced from the ore. The effects of these compositional differences on the leaching rates of glasses made from ores cannot be predicted. Sodium and potassium oxides are most readily leached from glasses⁷ and glass durability is often measured by changes in their

concentrations. However, a high silica content also results in increased glass durability.

If radioactive ions are dissolved in glass or dispersed on a nearly molecular scale, they can be removed only by dissolving the silica matrix. The rate of dissolving would, in turn, be influenced to some extent by the concentration of silica in an aqueous solution. The solubility of

amorphous silica in water at 363 K is approximately 350 ppm,³⁴ which in experiment F would correspond to 0.61% glass dissolved. This should not, however, be considered a limiting value since both supersaturation and precipitation of amorphous silica can occur.

The dissolving rates of the glass in the present experiments were calculated with the assumption that the

Fig. 9. Cumulative dissolving rate of melt glass for Experiment F.

radioactive species that were counted had been uniformly distributed in the glass and had been leached at the same rates as all the other components. All information on these rates are in general agreement. The data of experiment F is the most useful, since it covers the longest time and shows the least scatter. Figure 9 shows a relatively high early rate, declining over a six-month period to an approximately constant rate of about 1×10^{-10} kg/s·m². The reasons for such a change in dissolution rate are not known. In addition to the possibilities discussed previously, the high initial rate may reflect high surface ener-

gies of the freshly broken glass particles or even dissolution of dust-size glass particles in the final glass-size fraction, which was screened but not washed.

Data on the dissolving rates of closely comparable glasses under the conditions of these experiments are not available. However, similar rates have been observed for durable, high-silica, low-alkali glass in water and acid at this temperature.³⁵ As a practical matter, the long-term rate found should be useful in estimating the availability of radionuclides that have been incorporated in melted and resolidified rock in an actual nuclear mine.

Summary and Conclusions

Two important types of reactions of radionuclides in the nuclear chemical mining of copper were investigated. The first is the dissolving of fission and activation products from the nuclear chimney by leaching solutions. The second is the sorption of such dissolved species on the surfaces of minerals.

Leaching experiments showed that radioactive glass formed from melted rock is very effective for the fixation of radioactivity. It is corroded only slowly and uniformly by the acidic oxidizing solutions that

are generated in the pressure-leaching process.

Sorption reactions of radioactive tracers with ore and glass continue for a very long time, and the amount of activity on the solid phases usually continues to increase. A substantial part of the activity is sorbed by decomposition products of the ore, although the radionuclides studied differed in their behavior. In the case of ruthenium, antimony, cerium, and strontium, virtually all activity ended on solid products after three months of reaction, while

35, 70, and 90% of the cesium, europium, and yttrium, respectively, remained in solution. Low pH and high activities of other ions in solution reduced the sorption of ruthenium, antimony, cerium, cesium, and strontium but not europium and yttrium.

These results lead to some useful conclusions on the prospects for the

control of radioactive products in nuclear chemical mining. Leaching time and pH are variables that can be used, not only to optimize the copper recovery, but to control the radioactivity of the resulting leach solution. Under the conditions anticipated for an actual mine, a natural decontamination process also takes place.

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Appendix A: Computation of Dissolving Rates of Radioactive Melt Glasses

Let $A_a \equiv$ activity of ^{152}Eu in glass
 $A_b \equiv$ activity of ^{154}Eu in glass
 $A_c \equiv$ activity of ^{154}Eu from glass in solution
 $A_d \equiv$ activity of ^{152}Eu from glass in solution
 $A_e \equiv$ activity of ^{152}Eu , total, in solution
 $A_f \equiv$ activity of ^{152}Eu from tracer, in solution
 $A_g \equiv$ activity of ^{152}Eu from tracer, total
 $A_h \equiv$ activity of ^{152}Eu from tracer, sorbed
 $A_i \equiv$ activity of ^{152}Eu from glass, sorbed
 $A_j \equiv$ activity of ^{152}Eu from glass, total
 $A_k \equiv$ activity of ^{144}Ce in glass
 $A_l \equiv$ activity of ^{144}Ce in solution
 $A_m \equiv$ activity of ^{144}Ce , sorbed
 $A_n \equiv$ activity of ^{141}Ce , sorbed
 $A_o \equiv$ activity of ^{141}Ce in solution
 $A_p \equiv$ activity of ^{141}Ce , total
 $F \equiv$ fraction of glass dissolved
 $T_r \equiv$ run time, s
 $R \equiv$ mass/area ratio, 0.317 kg/m^2
 $D \equiv$ glass dissolving rate, $\text{kg/s}\cdot\text{m}^2$

The measured quantities were A_a , A_b , A_c , A_e , A_g , A_k , A_l , A_o , A_p , T_r , and R . Then

$$A_d = A_c \cdot A_a / A_b \quad (9)$$

$$A_f = A_e - A_d \quad (10)$$

$$A_h = A_g - A_f \quad (11)$$

$$A_i = A_d \cdot A_h / A_f \quad (12)$$

$$A_j = A_d + A_i \quad (13)$$

$$F_{\text{Eu}} = A_j / A_a \quad (14)$$

$$D_{\text{Eu}} = F_{\text{Eu}} \cdot R / T_r \quad (15)$$

Similar calculations were made using the activities of ^{155}Eu . The calculations using cerium species have the following steps:

$$A_n = A_p - A_o \quad (16)$$

$$A_m = A_1 \cdot A_n/A_o \quad (17)$$

$$F_{\text{Ce}} = (A_1 + A_m)/A_k \quad (18)$$

$$D_{\text{Ce}} = F_{\text{Ce}} \cdot R/T_r \quad (19)$$

In each experiment, activities of radioactive species in the original tracer solution, in the glass, and in the measured aliquots were corrected to a common reference time.

Appendix B: Data Tables

Table B1. Distribution coefficients K_{dm} for experiment A ($K_{da} = 0.3205 K_{dm}$).

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
0.054	3.899	5.397			3.954	0.258	18.114
9.961	74.211				21.463	0.762	100.198
21.091	133.489			4.461	33.163	0.877	
28.112				5.854	43.716	1.169	
35.120				6.839	49.137	1.390	
42.117				7.856	54.084	1.730	
48.094	221.432			8.304	54.546	1.903	

Table B2. Dissolving rate of ^{152}Eu from melt glass in experiment A.

Time (d)	Total percent nuclide dissolved	Glass- dissolving rate, ($\text{kg/s}\cdot\text{m}^2$)
21.091	0.0765	1.331×10^{-10}
28.112	.1772	2.312×10^{-10}
35.120	.1689	1.764×10^{-10}
42.117	.3249	2.830×10^{-10}
48.094	0.2626	2.003×10^{-10}

Table B3. Distribution coefficients K_{dm} for experiment B ($K_{da} = 0.0317 K_{dm}$).

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
6.987	1.748		4.552	0.431	2.732		6.308
18.782	5.894		19.954	20.643	3.115		
25.832	9.247		18.908	22.958	5.887		31.363
32.826			30.960	50.377	12.101		
40.193	18.912		32.498	18.768	10.962		41.818
46.982			22.631	18.476	15.043		

Table B4. Dissolving rate of ^{152}Eu from melt glass in experiment B.

Time (d)	Computation from $^{152}\text{Eu}/^{154}\text{Eu}$ in glass		Computation from $^{152}\text{Eu}/^{155}\text{Eu}$ in glass	
	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ⁻²)	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ⁻²)
6.987	0.0253	1.328×10^{-10}	0.0374	1.984×10^{-10}
18.782	.0894	1.746×10^{-10}		
25.832	.1298	1.843×10^{-10}	0.0788	1.119×10^{-10}
32.826	.2028	2.266×10^{-10}	.2670	2.984×10^{-10}
40.193	.1241	1.132×10^{-10}	.1767	1.613×10^{-10}
46.982	0.1384	1.081×10^{-10}	0.3181	2.484×10^{-10}

Table B5. Dissolving rate of ^{144}Ce from melt glass in experiment B.

Time (d)	Total percent nuclide dissolved	Glass dissolving rate ($\text{kg/s}\cdot\text{m}^2$)
6.987	0.0315	1.654×10^{-10}
18.782	.0711	1.389×10^{-10}
25.832	.0768	1.091×10^{-10}
32.826	.0903	1.009×10^{-10}
40.193	.1443	1.317×10^{-10}
46.982	0.1157	0.903×10^{-10}

Table B6. Distribution coefficients K_{dm} for experiment C ($K_{\text{da}} = 1.569 K_{\text{dm}}$).

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
0.945	0.318	1.589	0.282	0.198	0.772	0.345	0.689
9.837	2.982	10.142	3.957	0.710	3.382	0.468	8.717
14.872	4.177	15.300	5.211	0.961	4.061	0.623	13.661
20.723	5.932	27.616	7.812	1.067	5.183	0.763	23.619
28.678	9.486	73.563	12.828	1.243	6.958	0.741	58.330
35.682	15.786	187.908	19.032	1.517	7.658	0.848	145.774
41.702	19.966	241.974	24.302	1.439	7.007	0.877	156.642
49.639	31.305	465.543	31.766	1.837	6.657	1.086	
56.834	45.918	707.760	15.980	2.089	6.068	1.154	
62.851	53.632		45.096	1.829	5.241	0.683	
73.443	96.538	985.387	81.330	2.344	5.133	1.065	
77.849	105.993			2.497	5.039	1.171	
84.872				2.705	4.829	1.097	
91.817				3.382	5.360	1.340	
94.646 ^a	132.274		72.628	2.916	7.017	1.308	

^aData for cooled solution.

Table B7. Distribution coefficients K_{dm} for experiment D ($K_{da} = 1.569 K_{dm}$).

Time (d)	$^{103}_{Ru}$	$^{124}_{Sb}$	$^{141}_{Ce}$	$^{152}_{Eu}$	$^{134}_{Cs}$	$^{88}_{Y}$	$^{85}_{Sr}$
1.606	38.310	30.218	9.303	9.347	5.315	7.956	1.464
8.566		316.213	2.824	2.348	7.154	2.210	1.287
15.368			2.700	1.926	8.267	1.735	1.782
22.352			2.711	1.811	8.902	1.535	2.160
29.361			3.277	1.967	9.489	1.606	2.837
36.362			3.835	1.972	9.875	1.568	3.567
43.569			1.245	1.989	9.962	1.465	4.432
50.562			1.895	2.020	10.674	1.540	5.737
58.628			5.442	2.063	11.008	1.384	8.168
64.359			6.925	2.122	11.361	1.418	10.569
70.389			7.236	2.284	12.016	1.460	13.614
78.575			10.614	2.251	12.557	1.463	23.227
85.371			10.702	2.402	13.323	1.464	35.162
92.373			10.718	2.398	13.533	1.364	58.480
99.448			17.783	2.465	14.603	1.450	89.270
106.448			25.301	2.652	15.881	1.529	71.964
107.465	160.936	519.855	18.174	2.575	30.473	1.485	46.678

Table B8. The pH history of experiment D.

Time (d)	pH
0	5.25
1.606	3.60
8.566	2.85
15.368	2.55
22.352	2.45
29.361	2.34
36.362	2.20
43.569	2.13
50.563	2.12
64.359	1.83
70.389	1.73
78.575	1.71
85.371	1.65
92.373	1.62
99.448	1.55
106.478	1.57
107.465	1.98

Table B9. Distribution coefficients K_{dm} for experiment E ($K_{da} = 0.3205 K_{dm}$).

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
-2.125	0.071	1.526	0.223	0.081	3.029	0.110	0.297
2.858	5.125	22.740	15.323	0.623	18.420	0.300	20.119
5.976	9.071	54.946	69.575	0.978	45.916	0.433	240.029
9.841	8.031	42.042	20.875	1.134	44.176	0.514	35.292
16.653	14.431	81.052	44.970	1.261	62.341	0.539	70.342
19.668	24.518	185.212	101.523	1.738	90.946	0.778	342.831
24.670	29.277	165.190	54.300	2.340	85.606	0.886	319.963
26.910	30.086	233.839	50.056	3.495	55.579	1.227	159.636
32.709	43.881	216.441	50.934	3.925	71.548	1.352	420.266
40.698	41.534	343.661	35.387	5.033	41.026	2.177	123.929
45.837	53.531	439.546	32.287	5.295	36.482	2.410	91.447
52.879	52.106	444.766	33.049	5.570	23.299	2.583	115.145
60.858	59.546	392.837	34.249	6.465	14.506	3.092	94.391
68.732	80.435	406.419	39.431	7.176	13.269	3.968	114.079
74.880	108.730	1715.980	47.258	31.777	21.822	21.097	67.965
82.870	30.123	477.557	14.863	9.305	3.291	6.268	22.399
89.696	39.421	705.564	18.894	9.953	3.399	7.112	31.070
96.545	46.130	512.614	21.970	9.094	2.884	6.536	43.607
103.766	54.692	610.598	26.497	9.432	2.982	6.352	71.896
110.608	77.202	718.654	33.795	11.924	3.216	7.801	106.808
117.611	84.413	928.717	34.004	12.117	3.001	8.442	133.611
124.604	110.026	837.843	41.857	13.783	2.821	8.951	214.516
131.625	122.141	1153.632	52.413	16.599	2.730	10.403	305.599
138.625	82.439	1421.486	39.026	18.883	2.924	11.963	105.287
145.644	99.008	1783.030	36.685	17.499	2.907	12.010	113.087
152.644	138.383	1704.276	46.451	22.340	3.011	14.499	173.577
159.678	116.715	1605.167	32.267	24.071	3.165	16.148	82.084
166.594	70.523	1461.596	20.935	21.105	2.931	15.938	68.000
173.639	113.092	2049.000	24.711	23.192	3.111		85.216
180.604	190.085	1779.651	31.750	30.574	3.113	21.416	133.086
187.604	194.489	1670.265	37.182	29.133	2.796	24.481	265.712
194.604	128.064	2050.970	36.163	38.643	3.074	25.667	145.784

Table B9 (continued)

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
201.604	136.273	1585.291	31.151	35.225	3.122	28.760	97.226
211.907	103.259		30.975	40.522	3.947	34.744	75.602
215.604	127.972	2483.072	29.078	33.300	3.198	31.585	75.717
222.584			32.440	35.110	2.868	30.535	112.601
228.625		2471.394	40.396	19.421	3.569	16.368	133.204
236.644	95.572		34.694	23.012	3.256	17.695	140.109
243.644		2419.047	39.468	23.099	3.163	18.876	131.095
250.625			47.719	21.418	3.261	20.875	81.276
255.625			28.057	16.261	4.769	19.129	82.252

Table B10. The pH history of experiment E.

Time (d)	pH	Time (d)	pH
-2.877	0.950		
-2.125	1.340		
2.858	1.550	138.625	1.000
5.976	1.322	145.644	1.100
9.841	1.366	152.644	1.070
16.653	1.195	159.678	1.060
19.668	1.260	166.594	1.100
24.670	1.203	173.639	1.110
26.910	0.935	180.604	1.070
32.709	1.375	187.604	1.045
40.698	0.675	194.604	1.070
45.837	1.130	201.604	1.110
52.879	1.022	211.907	1.190
60.858	0.895	215.604	1.130
68.732	1.145	222.584	1.170
74.880	1.325	228.625	1.180
82.870	0.571	236.644	1.080
89.696	0.935	243.644	1.110
96.545	1.070	250.625	1.110
103.766	1.160	255.625	1.145
110.608	1.000		
117.611	1.085		
124.604	1.000		
131.625	0.820		

Table B11. Distribution coefficients K_{dm} for experiment F ($K_{da} = 0.0317 K_{dm}$).

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Sr
-1.179	0.331	2.918	0.298		3.044	0.123	0.250
2.990	1.782	515.375	0.687		3.008	0.064	0.830
6.045	2.810	867.735	1.748	1.153	3.307	0.566	2.175
9.649	4.090	1043.613	2.603	1.444	3.777	1.036	3.133
16.794	6.180		3.933	2.536	4.463	2.227	4.788
22.629	7.003	827.613	4.320	2.911	4.554	2.572	5.821
30.646	7.934	808.521	5.816	4.313	4.808	3.116	7.774
37.670	9.190	918.478	7.020	4.494	5.192	3.923	9.900
44.719	10.084	1147.383	10.038	7.332	5.149	4.783	14.638
52.653	12.024	1056.857	12.885	9.748	5.973	6.903	20.753
59.823	12.199	1180.216	14.309	10.196	5.932	7.137	22.634
67.694	13.805	1022.552	16.698	12.244	6.312	7.543	31.910
75.677	16.396	1503.235	23.248	18.233	7.683	10.803	39.171
82.653	17.071	1741.933	28.510	21.313	8.117	12.764	51.787
96.506	23.493		42.860	41.429	11.983	21.544	88.854
103.724	29.254		56.284	45.232	14.085	24.138	110.796
110.760	28.816		47.398	47.243	13.092	24.018	138.654
117.764	31.730		68.443	51.125	15.015	29.600	171.490
124.761	31.492		65.882	54.888	13.611	30.250	139.389
131.781	32.788		56.102	55.734	13.474	30.675	191.650
138.810	31.664		98.572	58.297	13.819	32.426	194.089
145.642	36.573		66.911	72.482	16.729	39.611	245.387
152.642	42.928		102.624	86.394	17.555	42.958	224.752
159.769	47.562		117.155	97.050	20.569	48.241	311.585
166.615	53.755			110.331	23.498	57.550	
173.615	62.602		139.334	108.866	22.479		499.623
180.615	62.296			114.897	22.411	50.941	
187.615	79.007			116.281	24.144	52.897	
194.615	49.275			139.776	26.312	73.963	
201.615	39.483			142.015	28.380	60.330	
211.594	50.318	483.752		137.689	27.133	73.386	
215.615	130.260			164.055	33.595	79.622	

Table B11 (continued)

Time (d)	^{103}Ru	^{124}Sb	^{141}Ce	^{152}Eu	^{134}Cs	^{88}Y	^{85}Kr
222.594				162.194	38.996	216.730	
228.635	83.115			194.739	35.528	81.287	
236.625				181.998	36.796	101.933	
243.625	81.188			240.791	39.103	112.968	
250.625				285.512	44.306	133.174	

Table B12. Dissolving rate of ^{152}Eu from melt glass in experiment F.

Time (d)	Computation from $^{152}\text{Eu}/^{154}\text{Eu}$ in glass		Computation from $^{152}\text{Eu}/^{155}\text{Eu}$ in glass	
	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ²)	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ²)
6.045	0.0409	2.482×10^{-10}		
9.649	.0529	2.011×10^{-10}		
16.794	.0736	1.608×10^{-10}		
22.629	.0901	1.461×10^{-10}		
30.646	.1065	1.275×10^{-10}	0.0784	0.938×10^{-10}
37.670	.1202	1.171×10^{-10}	.1419	1.382×10^{-10}
44.719	.1503	1.233×10^{-10}	.1669	1.369×10^{-10}
52.653	.1693	1.180×10^{-10}	.2084	1.452×10^{-10}
59.823	.1828	1.121×10^{-10}	.1606	0.985×10^{-10}
67.694	.1793	0.972×10^{-10}	.2539	1.376×10^{-10}
75.677	.2271	1.075×10^{-10}	.2603	1.262×10^{-10}
82.653	.2216	0.984×10^{-10}	.2248	0.998×10^{-10}
96.506	.2819	1.072×10^{-10}	.4039	1.535×10^{-10}
103.724	.2874	1.016×10^{-10}	.2748	0.972×10^{-10}
110.760	.3101	1.027×10^{-10}	.4684	1.551×10^{-10}
117.764	.2649	0.825×10^{-10}	.2803	0.873×10^{-10}
124.761	.3230	$.950 \times 10^{-10}$	.4299	1.264×10^{-10}
131.781	.3281	$.913 \times 10^{-10}$	.3403	0.947×10^{-10}
138.810	.3321	$.878 \times 10^{-10}$	.5248	1.387×10^{-10}
145.642	.3415	$.860 \times 10^{-10}$	.4791	1.207×10^{-10}
152.642	.3872	$.931 \times 10^{-10}$	.4308	1.035×10^{-10}
159.769	.4049	0.929×10^{-10}	.6199	1.423×10^{-10}
166.615	.4565	1.005×10^{-10}	.4548	1.001×10^{-10}
173.615	.4703	0.993×10^{-10}	.5096	1.077×10^{-10}
180.615	.4190	$.851 \times 10^{-10}$	.8959	1.820×10^{-10}
187.615	.4544	$.888 \times 10^{-10}$	.5111	0.999×10^{-10}
194.615	.4732	$.892 \times 10^{-10}$	.9078	1.711×10^{-10}
201.615	.4435	$.807 \times 10^{-10}$		
211.594	0.4858	0.842×10^{-10}	0.8230	1.427×10^{-10}

Table B12 (continued)

Time (d)	Computation from $^{152}\text{Eu}/^{154}\text{Eu}$ in glass		Computation from $^{152}\text{Eu}/^{155}\text{Eu}$ in glass	
	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ²)	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ²)
215.615	0.5026	0.858×10^{-10}	0.6280	1.068×10^{-10}
222.594	.4572	$.770 \times 10^{-10}$	.5461	0.900×10^{-10}
228.635	.5758	$.924 \times 10^{-10}$	.8152	1.308×10^{-10}
236.525	.5239	$.812 \times 10^{-10}$	.7066	1.095×10^{-10}
243.625	.5906	$.888 \times 10^{-10}$	.5222	0.786×10^{-10}
250.625	0.6574	0.961×10^{-10}	0.8408	1.229×10^{-10}

Table B13. Dissolving rate of ^{144}Ce
from melt glass in exper-
iment F.

Time (d)	Total percent nuclide dissolved	Glass- dissolving rate ₂ (kg/s·m ²)
30.646	0.1172	1.403×10^{-10}
37.670	.1300	1.266×10^{-10}
44.719	.1034	0.848×10^{-10}
59.823	.1552	$.952 \times 10^{-10}$
67.694	.1271	$.689 \times 10^{-10}$
152.642	0.2158	0.519×10^{-10}

Appendix C: Data Plots

Fig. C1. Ruthenium K_{da} on ore.

Fig. C2. Antimony K_{da} on ore.

Fig. C3. Cerium K_{da} on ore.

Fig. C4. Europium K_{da} on ore.

Fig. C5. Cesium K_{da} on ore.

Fig. C6. Yttrium K_{da} on ore.

Fig. C7. Strontium K_{da} on ore.

Fig. C8. Ruthenium K_{da} on glass.

Fig. C9. Antimony K_{da} on glass.

Fig. C10. Cerium K_{da} on glass.

Fig. C11. Europium K_{da} on glass.

Fig. C12. Cesium K_{da} on glass.

Fig. C13. Yttrium K_{da} on glass.

Fig. C14. Strontium K_{da} on glass.

Fig. C15. Cumulative dissolving rate of glass, Experiment A.

Fig. C16. Cumulative dissolving rate of glass, Experiment B.

Fig. C17. Percent of glass dissolved, Experiment F.

Fig. C18. The pH history of Experiment D.